# Uranium isotopic evidence for groundwater chemical evolution and flow patterns in the eastern Snake River Plain aquifer, Idaho

**Robert C. Roback***
*Environmental Division, M.S. J514, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

**Thomas M. Johnson**
*Department of Geology, University of Illinois at Urbana-Champaign, 245 Natural History Building, MC-102, Urbana, Illinois 61801, USA*

**Travis L. McLing**
*Idaho National Engineering and Environmental Laboratory, P.O. Box 1625, Idaho Falls, Idaho 83415, USA*

**Michael T. Murrell**
*Chemistry Division, M.S. J514, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

**Shangde Luo**
**Teh-Lung Ku**
*Department of Earth Sciences, University of Southern California, Los Angeles, California 90089-0740, USA*

## ABSTRACT

**The isotopic composition and concentration of uranium and strontium in groundwater, combined with solute concentration data, provide important details regarding groundwater geochemical evolution and flow-pathways in the eastern Snake River Plain aquifer. The study was conducted in the vicinity of the Idaho National Engineering and Environmental Laboratory (INEEL), Idaho, which has a long history of storing and disposing of radioactive waste, some of which has entered the aquifer.**

**Uranium concentrations in INEEL groundwater range from 0.3 to 3.6 ppb, and $^{234}U/^{238}U$ atomic ratios range between 0.000085 and 0.000168 (activity ratios of 1.5 to 3.1). All of the samples have natural $^{235}U/^{238}U$ ratios, and $^{236}U$ was not detected; thus, the trends delineated by the $^{234}U/^{238}U$ ratios reflect natural variations in the aquifer. Groundwater nearest the valleys that provides focused recharge to the Snake River Plain aquifer from the northwest has high $^{234}U/^{238}U$ ratios when compared to values of regional groundwater flowing southwestward in the aquifer. Mixing of these water masses can account for the intermediate uranium isotope ratios of some of the samples; however, water-rock interaction must also be invoked to account for the observed trends in isotopic data. Uranium and $^{87}Sr/^{86}Sr$ isotope ratios are positively correlated and define a trend toward isotope ratios of the aquifer host rock. These relations indicate that dissolution and/or ion exchange are more important than alpha recoil or selective leaching in controlling $^{234}U/^{238}U$ ratios. As a result, $^{234}U/^{238}U$ ratios decrease along flow pathways toward the secular equilibrium values of the aquifer host rock. Uranium and strontium isotopic modification can be explained by incongruent dissolution of the host basalt.**

**Lateral distributions of $^{234}U/^{238}U$ ratios indicate elongate zones of high $^{234}U/^{238}U$ ratios extending southward from the mouths of Birch Creek and the Little Lost River. These elongate zones are interpreted as preferential flow paths. Two isolated pockets of groundwater located in the central and western parts of the study area have lower $^{234}U/^{238}U$ ratios than the adjacent aquifer water. Both of these zones are interpreted to contain stagnant waters that are relatively isolated from flow in the regional aquifer due to lower permeability. Physical and chemical evidence strongly suggests that the stagnant zones are dominated by water from the Big Lost River that infiltrated via flood control ponds (spreading areas), playas, and the riverbed.**

**Keywords: flow, groundwater, Snake River plain, strontium, uranium disequilibrium.**

## INTRODUCTION

Radiogenic isotopes can provide a unique fingerprint of groundwater from different sources that, in many cases, is not provided by elemental solute concentration data. Isotopic disequilibrium between groundwater and the host rock is common, owing to the slow reaction kinetics in these low-temperature environments. The isotopic signature of groundwater commonly reflects conditions far from its present location. Natural isotope ratios can therefore provide information regarding the source of groundwater, mixing of different groundwater masses, flow patterns, and rates and progress of water-rock interaction (Johnson and DePaolo 1994, 1997). Isotopic studies may provide a clearer and more detailed picture of groundwater flow patterns than do other chemical and physical methods and thus represent important tools when establishing remediation efforts and aquifer management policy.

Groundwater flow in fractured aquifers is commonly characterized by heterogeneous flow velocities, residence times, and extents of

*E-mail: roback@lanl.gov.

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water-rock interaction. The difficulties in predicting such flow heterogeneity introduce large uncertainties in predicting contaminant transport and in groundwater management. In this study, we examine $^{234}U/^{238}U$ isotopic ratios in a freshwater, fracture-flow aquifer, the eastern Snake River Plain aquifer, and we interpret these results in conjunction with strontium isotope and solute concentration data. The results provide important information regarding groundwater flow patterns and geochemical evolution of the aquifer.

$^{234}U$ ($t_{1/2} = 2.48 \times 10^5$ yr) is part of the $^{238}U$ ($t_{1/2} = 4.47 \times 10^9$ yr) radioactive decay series. The $^{234}U/^{238}U$ ratio in rocks is generally close to the secular equilibrium value of approximately $55 \times 10^{-6}$ (55 ppm), equivalent to a $^{234}U/^{238}U$ activity ratio of 1. However, $^{234}U/^{238}U$ ratios in groundwater are typically greater than the secular equilibrium value because of preferential dissolution of $^{234}U$ from crystallographic defects created by alpha recoil and because of direct ejection of $^{234}Th$ (which decays in about 24 days to $^{234}U$) into groundwater by alpha recoil (see Gascoyne, 1992 for review and additional hypotheses). Natural variations in groundwater $^{234}U/^{238}U$ reflect the competing effects of decreased $^{234}U$ due to radioactive decay, addition of $^{234}U$ by selective leaching and alpha recoil, and addition of uranium with equilibrium $^{234}U/^{238}U$ isotopic ratios by rock and/or mineral dissolution. Removal of uranium from solution by precipitation or sorption does not affect the isotopic ratio. Uranium is generally quite soluble in oxidizing waters and is readily hydrolyzed to form uranyl complexes (Langmuir, 1978; Gascoyne, 1992, and references therein). Typical surface and groundwater contain dissolved uranium in the parts-per billion (ppb, $10^{-9}$) range. The high solubility of uranyl complexes provides a natural isotope tracer that is relatively conservative in oxidizing aquifers.

## HYDROGEOLOGIC SETTING

The eastern Snake River Plain aquifer (Fig. 1) is one of the largest aquifers in the United States and is the most widely used source of water for drinking and agriculture in southern Idaho. The aquifer encompasses approximately 25 000 km$^2$ and stores 1.2 to 2.5 $\times 10^{12}$ m$^3$ (1 to 2 billion acre-feet) of water (Lindholm and Vaccaro, 1988). Most of the recharge to the aquifer is from the Snake River and its tributaries on the northeastern and eastern edge of the aquifer. Also important in the vicinity of the Idaho National Engineering and Environmental Laboratory (INEEL) (Figs. 1 and 2) is recharge from the Birch Creek, Little Lost River, and Big Lost River valleys to the north and west of the site. Regional flow in the aquifer is from the northeast to the southwest. Groundwater is discharged in a series of large springs between Twin Falls and King Hill (Fig. 1). The combination of low precipitation and high evaporation rate results in negligible direct recharge via infiltration of local precipitation. However, infiltrating waters from the Big Lost River, flood control basins, playas, and Mud Lake (Fig. 2B) may constitute a volumetrically significant percentage of the aquifer water in local areas.

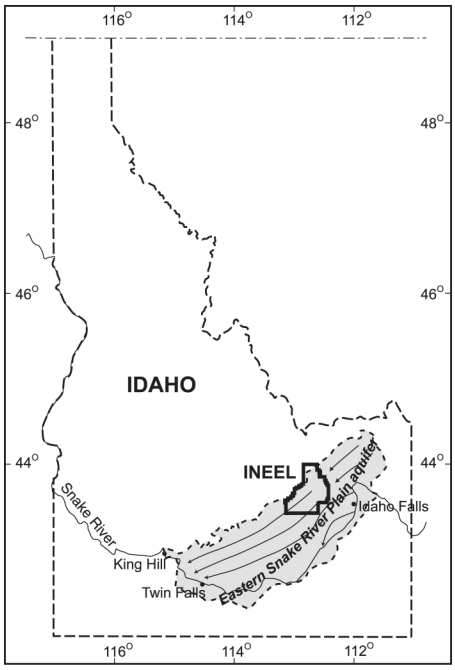


**Figure 1. Map of Idaho showing the eastern Snake River Plain aquifer and the location of the INEEL. Arrows show principal groundwater flow directions.**

The eastern Snake River Plain aquifer consists of a thick sequence of Quaternary and Tertiary basalt and lesser amounts of interlayered sedimentary rocks. The aquifer is unconfined and flow is fracture dominated. Hydraulic conductivity in the aquifer varies over several orders of magnitude (Lindholm and Vaccaro, 1988); the highest values are in highly fractured rubble zones that typify the tops and bottoms of individual basalt flows. Depth to the water table is approximately 60 m near the northern boundary of the INEEL and approximately 200 m near the southern boundary.

Groundwater in the Snake River Plain aquifer is a calcium–sodium-bicarbonate type. The groundwater typically contains <325 mg/L total dissolved solids, has a pH between 7 and 9, has temperatures ranging from 10 and 18 °C, and is saturated in dissolved oxygen (Wood and Low, 1986). Under these conditions, uranium is expected to be quite soluble and will most likely be present as uranyl carbonate complexes (Langmuir, 1978).

INEEL is a Department of Energy facility that encompasses approximately 2300 km$^2$ in the west-central part of the eastern Snake River Plain (Figs. 1 and 2). From the early 1950s, to the early 1980s, low-level radioactive and nonradioactive waste was disposed of via injection wells that penetrate to the aquifer. Contaminant plumes extend down gradient from these injection wells, and some contaminants can be tracked to near the southern boundary of the site (e.g., Bartholomay et al., 1995). In addition, there are several surface and shallowly buried waste storage sites that have leaked contaminants to the environment. Concerns about existing contamination and the potential for more have prompted numerous studies to better understand groundwater flow and contaminant migration in the aquifer (e.g., Welhan and Reed, 1997; Beasley, et al., 1998; Luo et al., 2000; Johnson et al., 2000; this study).

## SAMPLING AND ANALYTICAL TECHNIQUES

Sixty-six groundwater samples, two river samples and one hot-spring sample collected within or near the INEEL were analyzed for uranium isotopic composition and concentration (Table 1 and Fig. 2). Most of these samples were also analyzed for strontium isotopic composition and concentration as well as major cations (Johnson et al., 2000). Thirteen samples were collected on two separate occasions spanning up to 27 months between sampling to test for short-term variability. All groundwater samples are from the upper part of the basalt aquifer, with the exception of P&W 2, which is from a perched water zone in the basalt, and INEL-1, from which two samples from depths of approximately 610 and 3139 m (2000 and 10 300 ft) were analyzed.

Prior to sample collection, all wells were purged approximately three well-bore volumes to minimize effects of aging and well construction. Samples were filtered to 0.45 μm and acidified in the field to pH <2 with high-purity nitric acid. All samples were collected in acid-washed HDPE bottles. Uranium concentration and isotopic composition were determined by isotope-dilution thermal ionization mass spectrometry. Sample preparation

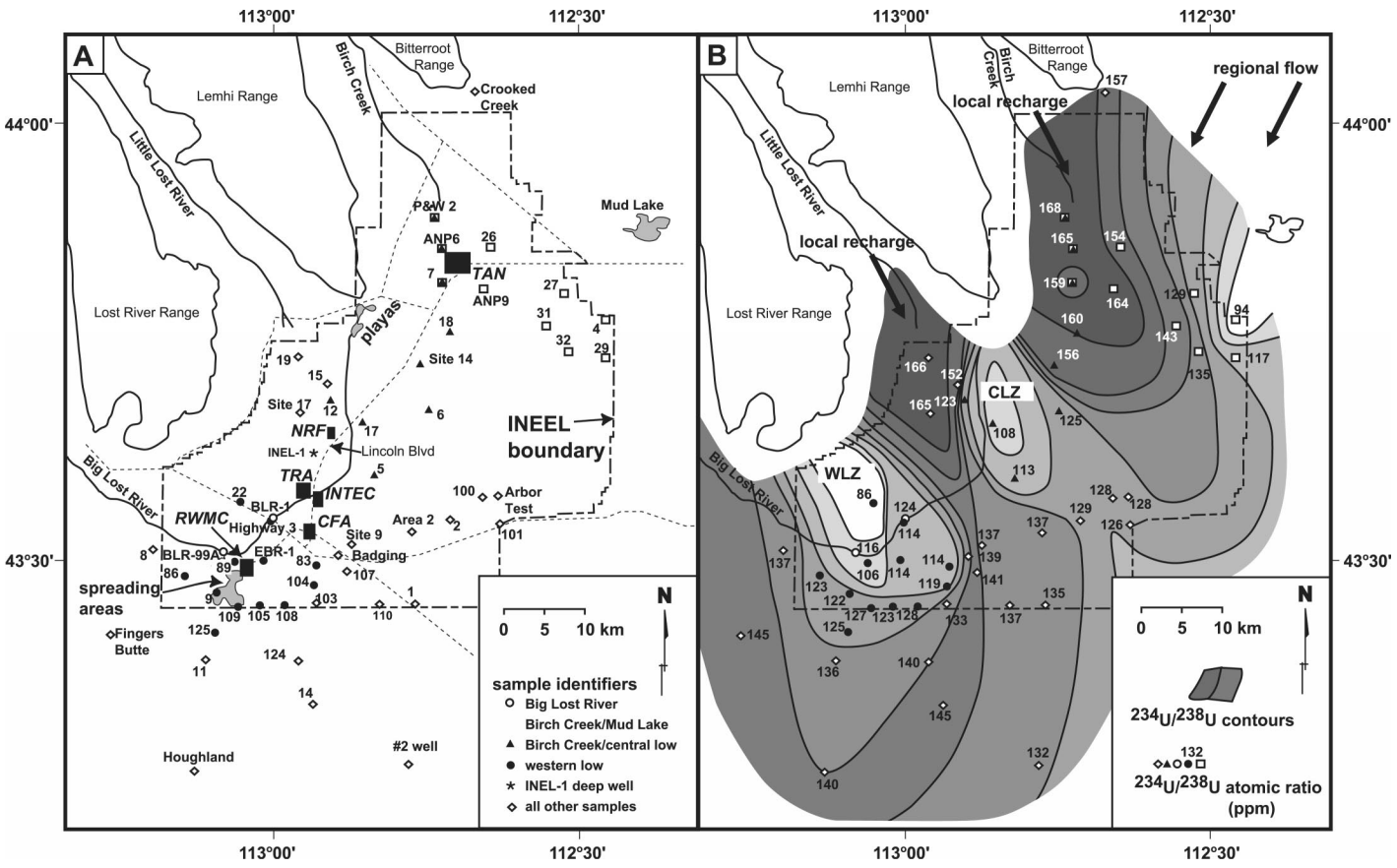


**Figure 2. Maps showing INEEL and vicinity. A: Important features and well identifiers for samples used in this study. Wells labeled only as numbers are U.S. Geological Survey (USGS) wells. Symbols for sample groupings (discussed in the text) are also used in Figures 3 and 4. B: Measured $^{234}U/^{238}U$ atomic ratios in parts per million (ppm) and a hand contour of those values for all aquifer samples. Kriging of data yields very similar contour patterns. The contours in the northeast corner of the study area are interpreted on the basis of solute concentration data, which generally show distinct concentric patterns with high values centered on Mud Lake (Johnson et al., 2000). Contour intervals are >160, 160–150, 150–140, 140–130, 130–120, 120–110, 110–100, <100. CLZ and WLZ are central and western low-isotope-ratio zones. Sturm–1 and Condie Hot Spring are outside the map area.**

and analytical procedures are discussed further in Appendix 1. Field blanks consisted of ultrapure water (with a known uranium concentration) that was transported to the field in precleaned Teflon bottles and then handled and analyzed in the same manner as a typical sample. All field blanks had comparable values to laboratory procedural blanks, which ranged from 5 to 50 pg. The maximum blank correction to the uranium concentration for all samples is less than 0.3%.

## RESULTS

Uranium concentrations in 68 of the 70 groundwater samples reported in Table 1 range from 0.33 to 3.59 ppb, values that are typical for oxidizing groundwater (Osmond and Cowart, 1992). Uranium concentrations do not vary systematically across the study area, nor are they correlated with major cation concentrations or ratios. Samples representing water from greater depths (INEL-1, 3139 m [10 300 ft] and Condie Hot Spring) have smaller uranium concentrations (0.19 and 0.03 ppb, respectively) than do shallower samples. These relationships are consistent with lesser solubility of uranium under the more reducing conditions expected at depth.

The INEEL has a long history of disposing of isotopically modified uranium, and uranium with non-natural isotopic composition has been detected in the aquifer down gradient from the Idaho Nuclear Technology and Engineering Center (INTEC) (Fig. 2A) deep disposal well (Beasley et al., 1998). Although most of the samples analyzed in this study were collected far from known contamination sites, a few of the samples are from areas that may contain non-natural uranium. For this reason, all samples from wells that are within or near the contaminant plume extending south of the INTEC (e.g., Beasley et al., 1998), as well as some of the samples from other wells, were examined for the presence of anthropogenic $^{236}U$ and non-natural $^{235}U/^{238}U$, neither of which was detected. These data demonstrate that the measured $^{234}U/^{238}U$ isotopic ratios represent natural values.

Most $^{234}U/^{238}U$ atomic ratios range between 0.000085 and 0.000173 (85 to 173 ppm, Table 1)[1]. The only exception is Condie Hot Spring, which has a ratio of 0.000292. Short-term variability in uranium isotopic composition as determined by analysis of duplicate samples col-

[1]In this report $^{234}U/^{238}U$ ratios are reported as atomic ratios rather than activity ratios and are stated as ppm (e.g. $0.000165 = 165 \times 10^{-6} = 165$ ppm). To convert to activity ratios, the atomic ratio is divided by $\lambda_{238}/\lambda_{234}$, a value of about 0.000055 (55 ppm). Thus, most $^{234}U/^{238}U$ activity ratios reported in this study are between 1.5 and 3.1.

TABLE 1. URANIUM CONCENTRATION AND ISOTOPIC COMPOSITION

| Sample | Date collected | Latitude (°N) | Longitude (°W) | Total penetration below water table (m) | Screen thickness below water table (m) | Sample weight (g) | U (ppb) | ± (%)* | $^{234}U/^{238}U$ (ppm) | ± (%)* |
|---|---|---|---|---|---|---|---|---|---|---|
| USGS 1 | 4/21/97 | 43.4500 | 112.7856 | 14.0 | 9.1 | 75.4 | 1.70 | 0.32 | 134.6 | 0.39 |
| USGS 1 LVS† | 8/31/98 | 43.4500 | 112.7856 | | | 99.4 | 1.72 | 0.24 | 134.0 | 0.14 |
| USGS 1 LVS duplicate | 8/31/98 | 43.4500 | 112.7856 | | | 179.5 | 1.72 | 0.23 | 133.8 | 0.21 |
| USGS 2 LVS | 8/26/98 | 43.5556 | 112.7231 | 14.3 | 6.4 | 111.7 | 1.82 | 0.22 | 129.0 | 0.13 |
| USGS 4 | 4/28/97 | 43.7825 | 112.4728 | 88.7 | 81.7 | 58.9 | 3.59 | 0.27 | 94.3 | 0.33 |
| USGS 5 | 10/10/96 | 43.5950 | 112.8269 | 10.4 | 6.7 | 105.2 | 1.50 | 0.22 | 112.5 | 0.13 |
| USGS 6 | 7/16/96 | 43.6753 | 112.7603 | 63.1 | 26.8 | 10.6 | 1.75 | 0.13 | 124.7 | 0.18 |
| USGS 6 LVS | 9/12/97 | 43.6753 | 112.7603 | | | 89.7 | 1.75 | 0.21 | 125.1 | 0.16 |
| USGS 7 | 4/21/97 | 43.8208 | 112.7442 | 302.1 | 41.1 | 94.9 | 2.26 | 0.23 | 158.9 | 0.12 |
| USGS 8 | 10/8/96 | 43.5225 | 113.1994 | 14.0 | 9.1 | 193.3 | 2.10 | 0.20 | 136.7 | 0.12 |
| USGS 9 LVS | 9/1/98 | 43.4590 | 113.0777 | 8.5 | 8.5 | 166.9 | 1.57 | 0.21 | 122.0 | 0.12 |
| USGS 11 | 10/9/96 | 43.3933 | 113.1117 | 30.5 | 9.1 | 98.6 | 1.67 | 0.22 | 135.8 | 0.12 |
| USGS 12 LVS | 9/10/97 | 43.6906 | 112.9186 | 111.9 | 32.0 | 72.2 | 2.36 | 0.26 | 123.0 | 0.21 |
| USGS 14 | 4/10/97 | 43.3386 | 112.9422 | 11.0 | 11.0 | 104.5 | 2.37 | 0.21 | 145.1 | 0.15 |
| USGS 15 | 7/25/96 | 43.7094 | 112.9214 | 89.6 | 21.3 | 163.7 | 1.78 | 0.25 | 152.4 | 0.17 |
| USGS 17 | 10/16/96 | 43.6600 | 112.8650 | 44.2 | 29.9 | 47.9 | 1.71 | 0.34 | 107.8 | 0.46 |
| USGS 17 LVS | 9/11/97 | 43.6600 | 112.8650 | | | 91.3 | 1.75 | 0.21 | 108.0 | 0.15 |
| USGS 18 | 7/19/96 | 43.7611 | 112.7358 | 18.3 | 7.3 | 12.3 | 2.06 | 0.31 | 159.8 | 0.12 |
| USGS 18 LVS | 9/12/97 | 43.7611 | 112.7358 | | | 90.1 | 2.07 | 0.21 | 160.1 | 0.13 |
| USGS 19 | 10/15/96 | 43.7406 | 112.9656 | 38.7 | 6.4 | 50.0 | 1.56 | 0.21 | 165.4 | 0.14 |
| USGS 19 LVS | 9/10/97 | 43.7406 | 112.9656 | | | 84.3 | 1.59 | 0.21 | 166.1 | 0.12 |
| USGS 22 | 7/18/96 | 43.5728 | 113.0547 | 14.0 | 4.6 | 46.7 | 0.33 | 0.27 | 84.9 | 0.86 |
| USGS 22 LVS | 8/31/98 | 43.5728 | 113.0547 | | | 535.5 | 0.43 | 0.22 | 85.9 | 0.16 |
| USGS 26 | 10/15/96 | 43.8694 | 112.5383 | 18.3 | 10.4 | 46.5 | 2.39 | 0.36 | 155.1 | 0.40 |
| USGS 26 LVS | 9/3/98 | 43.8694 | 112.5383 | | | 74.5 | 2.41 | 0.29 | 154.0 | 0.18 |
| USGS 27 | 10/15/96 | 43.8142 | 112.6000 | 26.2 | 6.1 | 45.7 | 3.04 | 0.19 | 129.2 | 0.10 |
| USGS 27 LVS | 9/2/98 | 43.8142 | 112.6000 | | | 89.3 | 3.10 | 0.21 | 129.4 | 0.13 |
| USGS 29 | 7/19/96 | 43.7353 | 112.4808 | 21.6 | 18.9 | 11.4 | 2.00 | 0.21 | 117.2 | 0.24 |
| USGS 31 LVS | 9/2/98 | 43.7736 | 112.5414 | 54.0 | 43.6 | 85.2 | 2.27 | 0.22 | 143.4 | 0.17 |
| USGS 32 | 7/19/96 | 43.7456 | 112.5392 | 31.1 | 26.2 | 41.4 | 2.53 | 0.16 | 134.7 | 0.19 |
| USGS 83 LVS | 8/28/98 | 43.5064 | 112.9376 | 78.3 | 71.9 | 100.7 | 1.37 | 0.23 | 113.6 | 0.20 |
| USGS 86 | 10/11/96 | 43.4928 | 113.1336 | 14.0 | 14.0 | 170.4 | 1.03 | 0.20 | 122.9 | 0.13 |
| USGS 86 LVS | 9/8/97 | 43.4928 | 113.1336 | | | 105.0 | 0.99 | 0.88 | 122.3 | 0.66 |
| USGS 89 | 7/17/96 | 43.5014 | 113.0586 | 17.1 | 17.1 | 475.6 | 1.16 | 0.27 | 105.5 | 0.30 |
| USGS 100 | 4/28/97 | 43.5842 | 112.6683 | 24.1 | 24.1 | 147.2 | 1.50 | 0.25 | 127.8 | 0.13 |
| USGA 101 | 10/10/96 | 43.5486 | 112.6386 | 29.0 | 29.0 | 71.1 | 1.32 | 0.21 | 125.4 | 0.16 |
| USGS 101 LVS | 9/8/97 | 43.5486 | 112.6386 | | | 94.8 | 1.36 | 0.25 | 125.7 | 0.18 |
| USGS 103 | 7/15/96 | 43.4539 | 112.9353 | 55.2 | 55.2 | 11.2 | 1.49 | 0.11 | 132.5 | 0.18 |
| USGS 103 LVS | 9/9/97 | 43.4539 | 112.9353 | | | 76.6 | 1.46 | 0.51 | 133.1 | 0.23 |
| USGS 104A | 7/15/96 | 43.4822 | 112.9356 | 45.1 | 45.1 | 12.2 | 1.52 | 0.12 | 118.9 | 0.20 |
| USGS 105 | 5/5/97 | 43.4508 | 113.0050 | 40.8 | 40.8 | 237.6 | 2.16 | 0.22 | 122.6 | 0.20 |
| USGS 107 LVS | 8/27/98 | 43.4950 | 112.8910 | 65.2 | 65.2 | 93.7 | 2.20 | 0.24 | 140.9 | 0.17 |
| USGS 108 LVS | 9/9/97 | 43.4497 | 112.9739 | 47.5 | 47.5 | 71.8 | 1.76 | 0.21 | 127.7 | 0.19 |
| USGS 109 LVS | 9/1/98 | 43.4503 | 113.0488 | 55.5 | 55.5 | 199.9 | 1.73 | 0.21 | 127.0 | 0.13 |
| USGS 110 LVS | 8/25/98 | 43.4547 | 112.8375 | 65.2 | 61.0 | 106.8 | 2.03 | 0.21 | 137.0 | 0.12 |
| USGS 124 | 4/10/97 | 43.3853 | 112.9753 | 35.1 | 15.2 | 115.8 | 1.61 | 0.21 | 140.3 | 0.13 |
| USGS 125 | 10/11/96 | 43.4339 | 113.0911 | 43.6 | 43.6 | 94.8 | 2.17 | 0.20 | 124.7 | 0.14 |
| ANP 9 | 4/28/97 | 43.3853 | 112.9753 | 98.1 | 23.5 | 52.1 | 2.37 | 0.19 | 164.4 | 0.13 |
| ANP 6 | 7/19/96 | 43.8644 | 112.7419 | 27.7 | 22.9 | 41.3 | 2.12 | 0.13 | 165.4 | 0.18 |
| ANP 6 LVS | 9/11/97 | 43.8644 | 112.7419 | | | 89.9 | 2.25 | 0.41 | 165.1 | 0.22 |
| ArborTest1 | 10/10/96 | 43.5856 | 112.6467 | 35.7 | 30.5 | 73.4 | 1.69 | 0.20 | 128.2 | 0.11 |
| AREA 2 | 7/18/96 | 43.5397 | 112.7839 | 65.5 | 48.5 | 10.3 | 1.94 | 0.16 | 136.6 | 0.17 |
| Badging Facility | 7/17/96 | 43.5117 | 112.8975 | 47.2 | 4.0 | 145.7 | 1.50 | 0.25 | 139.2 | 0.32 |
| Big Lost River 1 | 4/10/97 | 43.5493 | 113.0069 | SS | | 86.1 | 2.26 | 0.22 | 123.8 | 0.14 |
| Big Lost River 99A | 7/14/99 | 43.5158 | 113.0811 | SS | | 326.5 | 2.20 | 0.22 | 116.4 | 0.21 |
| Condie Hot Spring | 7/16/96 | 43.3342 | 113.9261 | SS | | 466.8 | 0.03 | 0.32 | 292.0 | 0.50 |
| Crooked Creek | 7/23/97 | 44.1742 | 112.6442 | N.A. | N.A. | 102.9 | 3.17 | 0.23 | 157.1 | 0.12 |
| EBR-1 | 10/16/96 | 43.5142 | 113.0072 | 146.0 | 144.8 | 143.9 | 2.03 | 0.21 | 113.7 | 0.18 |
| Fingers Butte | 6/3/97 | 43.3503 | 113.2853 | 20.4 | 15.8 | 160.5 | 1.78 | 0.22 | 144.7 | 0.13 |
| Highway 3 | 4/29/97 | 43.5492 | 113.0067 | 64.6 | 30.5 | 159.7 | 1.89 | 0.26 | 114.1 | 0.30 |
| Houghland | 6/3/97 | 43.2581 | 113.1169 | N.A. | N.A. | 160.6 | 1.81 | 0.21 | 139.8 | 0.12 |
| INEL-1 2000' | 3/24/79 | 43.6214 | 112.9431 | Sampled at 2000' during drilling | | 64.7 | 1.88 | 0.19 | 172.7 | 0.19 |
| INEL-1 10300' | 7/20/79 | 43.6214 | 112.9431 | Sampled at 10 300' during drilling | | 63.6 | 0.19 | 0.10 | 114.0 | 0.50 |
| P and W 2 | 10/15/96 | 43.9053 | 112.7586 | 23.5 | 20.7 | 42.1 | 1.86 | 0.22 | 167.8 | 0.12 |
| Site 9 | 7/22/96 | 43.5231 | 112.8836 | 178.0 | 113.7 | 11.2 | 1.68 | 0.19 | 137.4 | 0.17 |
| Site 14 | 8/20/96 | 43.7261 | 112.7753 | 138.4 | 55.5 | 45.5 | 2.07 | 0.20 | 155.8 | 0.11 |
| Site 14 LVS | 9/11/97 | 43.7261 | 112.7753 | | | 84.9 | 2.07 | 0.23 | 155.8 | 0.15 |
| Site 17 | 7/25/96 | 43.6742 | 112.9658 | 62.8 | 62.8 | 10.5 | 1.43 | 0.10 | 165.3 | 0.14 |
| Sturm 1 | 5/21/97 | 44.0944 | 111.4356 | N.A. | N.A. | 177.7 | 0.93 | 0.24 | 166.0 | 0.16 |
| #2 Well | 6/6/97 | 43.2725 | 112.7889 | N.A. | N.A. | 102.6 | 1.69 | 0.72 | 132.3 | 0.60 |

*Note*: SS—surface sample, N.A.—not available.
*Uncertainties are given at the 2-sigma level.
†LVS—large volume samples. Samples from these collections were also analyzed for short-lived U- and Th-series nuclides by Luo et al. (2000).

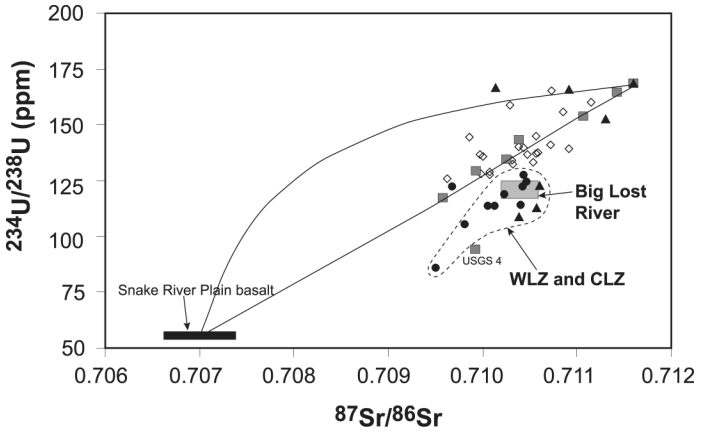


**Figure 3. Plot of $^{234}U/^{238}U$ versus $^{87}Sr/^{86}Sr$. The range of $^{87}Sr/^{86}Sr$ for typical Snake River Plain basalt is shown; the $^{234}U/^{238}U$ ratio is assumed to be near secular equilibrium. Curves show possible reaction paths discussed in text. Two-sigma error bars are smaller than symbols. See Figure 2 for explanation of symbols. $^{87}Sr/^{86}Sr$ data are from Johnson et al. (2000) and T. Bullen (1998, written commun.). Shaded rectangle shows the range of values for Big Lost River samples. CLZ and WLZ are central and western low-isotope-ratio zones.**

lected up to 27 months apart is insignificant (Table 1).

Unlike U concentrations, $^{234}U/^{238}U$ ratios show systematic lateral variations across the study area (Fig. 2B). Groundwater entering the aquifer from the Birch Creek, Little Lost River, and Big Lost River valleys to the north and west have high $^{234}U/^{238}U$ ratios relative to regional groundwater represented by samples in the eastern and southeastern parts of the INEEL. Water masses with these distinct isotopic signatures persist tens of kilometers away from their sources. For example, a narrow zone with high $^{234}U/^{238}U$ ratios extends southward from the mouth of the Little Lost River and remains well defined to the southern boundary of the INEEL. Two zones with relatively low $^{234}U/^{238}U$ ratios occur near the center and western parts of the INEEL.

## DISCUSSION

Natural variations in groundwater $^{234}U/^{238}U$ are affected by mixing of different groundwater masses, water-rock interaction, and, in very old water, decay of $^{234}U$. Groundwater residence times in the Snake River Plain aquifer (up to a few hundred years; Ackerman 1995; Luo et al., 2000) are very short relative to the half-life of $^{234}U$, thus eliminating radioactive decay as a factor in influencing $^{234}U/^{238}U$ ratios in this aquifer. Given the high solubility of uranyl complexes expected in this aquifer, uranium should act as a relatively conservative tracer of flow pathways and mixing. However, Luo et al. (2000) have shown that uranium is not perfectly conservative in this aquifer, and therefore the effects of water-rock interaction must also be taken into account. In the following sections we use uranium and strontium isotope ratios to delineate different sources of groundwater, to evaluate the roles of mixing and water-rock interaction, and to refine understanding of groundwater flow patterns in the aquifer.

### Groundwater Sources Delineated by Isotope Ratios

Regionally, the Snake River Plain aquifer flows from the northeast to the southwest beneath the INEEL. However, in the vicinity of the INEEL there are also several potential sources of recharge waters, including the Little Lost River and Birch Creek on the northwest edge of the aquifer, the Big Lost River and playas and flood-control basins along its course (Fig. 2A), and possibly upwelling groundwater. These recharge waters have uranium and strontium isotope ratios that are sufficiently distinct from those of the regional aquifer such that isotope ratios of these recharge waters are useful as natural tracers of groundwater flow.

A first-order observation of the isotopic data is that $^{234}U/^{238}U$ and $^{87}Sr/^{86}Sr$ ratios are higher in waters that emanate from the Little Lost River and Birch Creek valleys on the northwest edge of the aquifer than they are elsewhere in the aquifer (Fig. 2b; Johnson et al., 2000). In recent years, most of the surface water from the Little Lost River and Birch Creek valleys (Fig. 2) has been diverted for irrigation such that only small amounts reach the INEEL. Uranium and strontium isotope data are taken as evidence that these drainages do supply a significant volume of groundwater to the aquifer and thus constitute important recharge sources.

The Big Lost River intermittently flows westward onto the INEEL (Fig. 2) and rapidly infiltrates, via the riverbed, playas that mark its terminus and spreading areas to recharge the aquifer (Bennett, 1990). Since 1965, most of the flow from the Big Lost River has been diverted for flood control into spreading areas in the southwest part of the INEEL (Fig. 2A). Data from Bennett (1990) and the U.S. Geological Survey automated stream-flow system (U.S. Geological Survey, 1998) indicate that the total amount of water that infiltrated via the spreading areas and the riverbed south of Lincoln Boulevard (Fig. 2) between 1965 and 1998 is enough to replace the upper 21 m (70 ft) of aquifer over an area of approximately 417 $km^3$ (100 $mi^3$) (assuming 20% porosity, no water loss by evaporation). Infiltration of Big Lost River water results in a rapid rise and fall of water levels in wells near the river during periods of high and low river flows (Bennett, 1990). Thus, it is clear that the Big Lost River episodically contributes a significant volume of water to the aquifer.

Two areas with low $^{234}U/^{238}U$ (Fig. 2B) and $^{87}Sr/^{86}Sr$ (Johnson et al., 2000) isotope ratios lie near (but below) the terminal playas of the Big Lost River and the spreading centers, and thus may reflect infiltration of Big Lost River water. Johnson et al. (2000) termed these two areas the central low-isotope-ratio zone and the western low-isotope-ratio zone; we use that nomenclature in this paper. Uranium and strontium isotopic ratios from these samples define a distinct field in Figure 3 that plots below the trend defined by the rest of the aquifer samples with lower $^{234}U/^{238}U$ for corresponding $^{87}Sr/^{86}Sr$. The greatest uranium and strontium isotope ratios in both of these zones are nearly identical to those of the Big Lost River. Samples from the western low-isotope-ratio zone form an array that trends away from the isotopic composition of the Big Lost River toward U and Sr isotope ratios that are closer to those of the host basalt (Fig. 3), consistent with water-rock interaction as discussed below. These observations, combined with the physical evidence that the Big Lost River supplies a significant amount of water to the aquifer, argue that the two zones are dominated by infiltrated water from the Big Lost River.

Mud Lake (Fig. 2) is the terminus of a closed drainage basin that receives water from

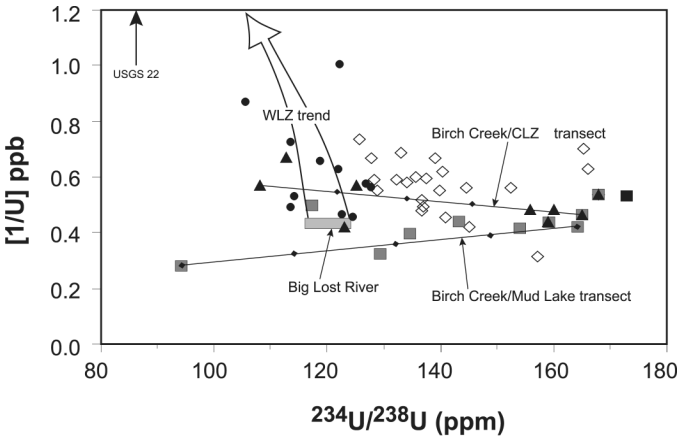


**Figure 4. Plot of reciprocal of uranium concentration versus $^{234}U/^{238}U$ atomic ratios. Possible mixing trends for the Birch Creek–Mud Lake and Birch Creek–central low-isotope-ratio zone, as well as the evolutionary trend for samples in the western low-isotope-ratio zone, are shown. Two-sigma error bars are smaller than symbols. Diamonds on mixing lines show 25% increments. See Figure 2 for explanation of symbols. Shaded rectangle shows the range of values for Big Lost River samples. CLZ and WLZ are central and western low-isotope-ratio zones.**

streams to the north and is a possible source of aquifer recharge via infiltration. High solute concentrations south of Mud Lake define tight concentration gradients on contour maps, reflecting the influence of significant volumes of infiltrated water derived from Mud Lake itself (Johnson et al., 2000; this study). In this study, sample USGS 4 is interpreted to contain significant proportions of water derived from Mud Lake, and uranium isotope contours (Fig. 2B) are drawn to reflect this.

Another potential source of recharge water in the aquifer is upwelling of water from depth; a mechanism that has been suggested to occur in both of the low-isotope-ratio zones (Roback et al., 1998; Smith and McLing, 1998). If upwelling waters did mix with the aquifer, the effect would most readily be noticed in the low-isotope-ratio zones, which are suspected to be regions of lower water flux relative to the rest of the aquifer (see below) and which have the lowest uranium concentrations of aquifer samples.

Careful examination of the uranium data suggests that upwelling thermal water does not cause the observed $^{234}U/^{238}U$ ratios of the low-isotope-ratio zones. Two samples representative of deeper water (INEL-1, 610 m [2000 ft] and Condie Hot Spring) have the highest $^{234}U/^{238}U$ ratios measured (172.9 and 292.0 ppm, respectively; Table 1), so input of similar water would drive the isotopic composition of water in the low-isotope-ratio zones toward higher $^{234}U/^{238}U$ values. One sample of deep water (INEL-1, 3139 m [10 300 ft]) has a $^{234}U/^{238}U$ ratio only slightly less than those of the Big Lost River, but still considerably greater than that of sample USGS 22. Furthermore, uranium concentrations in deeper waters are expected to be low, owing to the typical increase in reducing conditions with depth; this hypothesis is supported by the low uranium concentrations in the INEL-1 well at 3139 m [10 300 ft] depths and Condie Hot Spring (Table 1). Thus, mass balance considerations require large volumes of upwelling water relative to aquifer water in order to greatly influence the uranium isotope ratios anywhere in the aquifer. No physical (e.g., significantly elevated groundwater temperatures) or chemical (e.g., elevated solute concentrations similar to those of hot-spring samples [Johnson et al., 2000]) evidence supports this. Thus, the above considerations suggest that upwelling thermal waters do not constitute a significant proportion (more than a few percent) of the aquifer in the low-isotope-ratio zones (although small proportions are likely), nor is it likely that these waters greatly influence the uranium isotope ratios elsewhere in the aquifer.

### Origin of the Contrasting $^{234}U/^{238}U$ Ratios of the Recharge Valleys

The contrast in $^{234}U/^{238}U$ isotope ratios between waters emanating from the Little Lost River and Birch Creek valleys and the rest of the aquifer reflects the markedly different geology and hydrology of these recharge valleys when compared to those of the basalt aquifer. The recharge areas are in basin-and-range–type valleys that are filled with Quaternary gravel, sandstone, and shale of alluvial and fluvial origin (Kuntz et al., 1994). A significant component of groundwater flow in these recharge valleys is likely to be through intergranular pores. In contrast, the Snake River Plain aquifer consists dominantly of Quaternary basalt, and flow is fracture dominated. The clastic material that fills the recharge valleys is likely to have higher uranium concentrations (1 to 3 ppm for average sandstone and shale, respectively; Faure, 1986, Table 18.1) than does the basalt of the Snake River Plain aquifer (average of 0.37 ppm; Knobel et al., 1995). In addition, the clastic material of the recharge valleys contains a significant component of clastic material derived from old (Paleozoic to Precambrian) rocks, which core the mountain ranges that flank these recharge valleys, as indicated by high $^{87}Sr/^{86}Sr$ ratios of groundwater near these valleys (Johnson et al., 2000). In such old material, radiation damage to the crystal lattice of uranium-bearing minerals may enhance the effects of alpha recoil and selective leaching, resulting in larger $^{234}U$ excesses.

Thus, the marked difference in $^{234}U/^{238}U$ isotope ratios between the groundwater in the local recharge valleys and that in the basalt aquifer can be attributed to differences in flow mechanisms (porous vs. fracture flow), host rock lithology, and age of aquifer material. Alpha recoil and selective leaching result in enrichment of $^{234}U/^{238}U$ in groundwater flowing through clastic material in the recharge valleys to values of at least 160 to 168 ppm. $^{234}U/^{238}U$ ratios decrease away from these recharge valleys as a result of mixing with groundwater masses with lower $^{234}U/^{238}U$ ratios and/or release of uranium with near-equilibrium $^{234}U/^{238}U$ ratios from the basaltic host rock via dissolution as discussed below.

### Groundwater Mixing and $^{234}U/^{238}U$ Ratios

Given that the Snake River Plain aquifer in the vicinity of the INEEL comprises different water masses with distinct $^{234}U/^{238}U$ ratios and that uranium should behave as a relatively conservative tracer in this aquifer, uranium isotopes may be useful in determining mixing patterns and relative volumes. Mixing relations are manifested as linear trends on a plot of 1/U concentration vs. $^{234}U/^{238}U$ ratio (Faure, 1986). Uranium data for all groundwater samples from INEEL wells do not form a single collinear array (Fig. 4), because of the presence of multiple end-member waters with distinct uranium isotopic composition, concentration, and geographic distribution. The data set

was examined in greater detail by defining geographically based subsets designed to limit the number of potential end-member waters. The results are consistent with a strong influence of mixing across some of the mapped isotope gradients.

Mixing between groundwater from the local recharge valleys to the northwest and the groundwater of the regional aquifer to the east is to be expected and is evident from gradients in solute concentrations (Johnson et al., 2000). Samples along the transect extending from north of the Test Area North (TAN) to the eastern boundary of the INEEL south of Mud Lake (Fig. 2) display a crude linear trend (shaded squares in Fig. 4), suggesting that mixing between these two water masses is an important factor in the observed gradient in $^{234}U/^{238}U$ ratios. The mixing trend highlights the persistence of Birch Creek groundwater to a distance of about 10 km southeast of the TAN and then dilution by regional aquifer water near the site boundary. Deviations of the data from a perfectly linear array, however, indicate that two-component mixing alone does not explain the trend, and either multicomponent mixing or water-rock interaction must be invoked to explain modifications to the uranium data. The plot of $^{234}U/^{238}U$ vs. $^{87}Sr/^{86}Sr$ (Fig. 3) also shows a good correlation that is also consistent with two-component mixing. The exception is sample USGS 4, which is interpreted to contain significant proportions of recharge water derived from Mud Lake.

Uranium systematics along the transect from the mouth of Birch Creek to the central low-isotope-ratio zone also display a linear trend (black triangles in Fig. 4) consistent with mixing of two water masses. The most likely sources of the second mixing component are upwelling of deep groundwater and/or infiltration of Big Lost River surface water (see also Johnson et al., 2000). However, as discussed above, uranium isotopic data and mass balance considerations argue against large volumes of upwelling water in the low-isotope-ratio zones. It is more likely that the decrease in $^{234}U/^{238}U$ along this trend is produced by mixing of groundwater to the northeast with water that originated as surface water from the Big Lost River and was subsequently modified by water-rock interaction. This hypothesis is examined further below. No other linear trends in uranium systematics across isotope gradients are apparent (Fig. 4), probably because of multiple end-member mixing and/or water-rock interaction.

## Water-rock Interaction and Isotope Ratios

Uranium isotope-ratios are closely mimicked by $^{87}Sr/^{86}Sr$ ratios (Fig. 3). Groundwater from local recharge areas to the north and west has high $^{87}Sr/^{86}Sr$ ($>0.7110$), reflecting interaction with Paleozoic and Precambrian clastic rocks that core the mountain ranges (Kuntz et al., 1994) and/or clastic material derived from these old sources that fills the intervening valleys. The strontium isotope ratios of the groundwater decrease away from these areas (Johnson et al., 2000) and approach the isotopic composition of the host basalt ($^{87}Sr/^{86}Sr = 0.7070 \pm 0.0003$ [Leeman and Manton, 1971; Reed et al., 1997]). It is important to note that strontium isotope ratios are not influenced by alpha recoil or selective leaching, as are uranium isotope ratios. The good correlation displayed by these two isotopic systems indicates that dissolution of rock with near equilibrium $^{234}U/^{238}U$ ratios dominates over selective leaching and alpha recoil in controlling the $^{234}U/^{238}U$ ratios of the groundwater. $^{234}U/^{238}U$ and $^{87}Sr/^{86}Sr$ also define a trend toward the isotopic composition of the basalt (Fig. 3), by far the dominant rock type in the aquifer, which implies that U and Sr isotopics were modified through water-basalt dissolution.

The data were modeled by means of a two-end-member-mixing model (e.g., Faure, 1986; Banner and Hanson, 1990, Fig. 3). The endpoints selected for the model are average Snake River Plain basalt and average groundwater of the fluvial valleys to the northwest. The shape of the mixing line is determined by the Sr/U molar ratio of the end members. The upper hyperbola shows the mixing relation when the Sr/U molar ratio of average basalt (Knobel et al., 1995) and typical groundwater is used. This curve and others generated using the range of whole-rock Sr and U concentrations (Knobel et al., 1995) and typical values for INEEL groundwater do not fit the data. Thus, congruent dissolution of the bulk basalt cannot account for the observed correlation of isotope ratios. The lower hyperbola, which is a fit of the data points excluding the low-isotope-ratio zones, requires that the Sr/U molar ratio of the reactant is lower than that of bulk basalt. Basalt glass enriched in uranium and other incompatible elements is a reasonable candidate for the reactant. Glass is common in the basalt, constituting up to several percent of the rock (McLing, 1994), and it is less stable than crystalline components in the presence of water, readily reacting to form an outer hydrolyzed layer (Petit et al., 1990). Although the $^{87}Sr/^{86}Sr$ ratio in the glass is not known, deviations from that measured in the bulk basalt will be insignificant due to the young age (generally less than 1 m.y. [Anderson et al., 1997]) and the low $^{87}Rb/^{86}Sr$ ratios of the bulk basalt (Knoble et al., 1995). Finally, the Sr/U molar ratio of the glass, although not known for these rocks, is expected to be lower than that of the basalt, because of differences between Sr and U partitioning coefficients in mineral-melt systems.

The data are therefore interpreted to suggest that incongruent dissolution of basalt plays an important role in modifying the chemical and isotopic composition of groundwater in the aquifer. In such a scenario, uranium concentrations in the groundwater will increase until the solubility limit is reached. The data, however, show the opposite trend—i.e., uranium concentrations of the most reacted samples (e.g., USGS 22) have the lowest uranium concentration (Table 1 and Fig. 3). This implies that increasing water-rock interaction results in the release of uranium in secular equilibrium from the basalt by dissolution, and a removal of relatively greater amounts of uranium from solution by precipitation and/or ion exchange. Luo et al. (2000) reached a similar conclusion on the basis of uranium- and thorium-series disequilibria studies.

## Groundwater Flow Patterns

Although groundwater mixing and water-rock interaction affect the uranium and strontium isotope ratios of the Snake River Plain aquifer in this region, neither of these processes readily explains the observed spatial pattern of isotope ratios. Mixing of groundwater from the recharge valleys with water from the regional Snake River Plain aquifer to the east may partially explain the general northwest to southeast trend in uranium systematics and solute concentration data (Fig. 2; Johnson et al., 2000). Mixing, however, does not readily explain the shape or location of the zones with either high or low isotope ratios. It is also unlikely that isotopic heterogeneity in the host rock could impart the observed isotope-ratio pattern, because the chemical and isotopic composition of the host basalt is homogeneous on the scale of the study area (Leeman and Manton, 1971; Reed et al., 1997), and flow contacts (Anderson et al., 1996) track across $^{234}U/^{238}U$ and $^{87}Sr/^{86}Sr$ (Johnson et al., 2000) ratio contours. The observed isotopic pattern must therefore be due to spatial variations in groundwater flow.

We interpreted the two zones with high isotope ratios as preferential flow pathways along which groundwater from the Birch Creek and

Little Lost Rivers is channeled. Isotopic modification due to water-rock interaction or mixing is minimized along these zones because of short groundwater residence times and/or large volumes of water from the Birch Creek and Little Lost River basins entering the regional aquifer along narrowly focused zones. Conversely, the low-isotope-ratio zones are interpreted as zones of relatively stagnant groundwater that are physically isolated from the more rapidly flowing zones elsewhere in the aquifer by low-permeability zones. In the low-isotope-ratio zones, the combination of input from the Big Lost River and water-rock interaction result in low uranium and strontium isotope ratios. The relatively stagnant nature of these zones enhances the influence of infiltrating waters. Uranium and strontium isotope data do not appear to reflect the influence of infiltrated water where the Big Lost River crosses the zone with high isotope ratios in the central part of the INEEL, because the total groundwater flux is much greater relative to the infiltration flux in this preferential flow zone. Boundaries of the central and western low-isotope-ratio zones are well defined, indicating narrow mixing zones with the rest of the aquifer.

### Geologic Reason for Preferential Flow Pathways

The preferential flow pathways delineated by the isotope data are oriented south to southeast, at high angles to the general southwestward regional flow of the aquifer (e.g., Ackerman, 1995). High-transmissivity zones are typically located within rubbly rock that forms the tops and bases of basalt flows and within other features such as lava tubes and flow edges. Statistical analyses of transmissivity data in the aquifer (Welhan and Reed, 1997) predict directions of preferential hydraulic conductivity that are oriented north-northwest–south-southeast, identical to the orientation of flow pathways delineated in this study. Welhan and Reed (1997) suggested that the orientation of this hydraulic conductivity structure is related to the preferred alignment of volcanic features in the region. The study of Welhan and Reed (1997) successfully predicted the orientation of flow pathways as delineated by isotope data but their study did not predict the location of these pathways.

Flow channeling through primary volcanic features is a likely cause of the preferential pathways delineated by isotope data presented in this study and in Johnson et al. (2000). Groundwater that emanates from local recharge valleys of the Little Lost River and Birch Creek is funneled through high-transmissivity zones that correspond to statistically oriented primary volcanic structures in the Snake River Plain basalts. Channeling effects are most obvious nearest the recharge valleys, where volumes of the recharge groundwater are greatest. With increasing distance along flow pathways, these preferential pathways are deflected to more southward and southwestward directions and become more diffuse because of dilution and dispersion.

It is important to point out that hydraulic head data do not reveal the preferential flow pathways delineated in this study. Hydraulic head data reveal a general northeast-southwest–oriented gradient (Bartholomay et al., 1995), implying regional flow at nearly right angles to the preferential flow pathways identified here. This implies strong hydraulic anisotropy, in this case due to large variations in hydraulic conductivity in the fractured rock, which promotes preferential flow along pathways that exploit very slight head differentials.

## CONCLUSIONS

Radiogenic isotope ratios in groundwater provide powerful tools for understanding chemical and physical processes in aquifers. We have used natural variations in $^{234}U/^{238}U$ and $^{87}Sr/^{86}Sr$ ratios in groundwater from the Snake River Plain aquifer in the vicinity of the INEEL to identify chemically distinct water masses, to assess mixing and water-rock interaction, and to delineate detailed groundwater flow pathways. Mixing of groundwater from the mouth of Birch Creek with groundwater to the east and to the south appears to be an important process. Mixing of groundwater elsewhere, although potentially important, is difficult to document, because of complications caused by multiple end-member mixing and water-rock interaction. The uranium and strontium isotopic composition of groundwater evolves toward that of the host rock along flow pathways. Incongruent dissolution of the basalt must dominate over alpha recoil or selective leaching in the aquifer and can explain the observed isotopic modification.

Two preferential flow pathways extending southeast from the mouths of Birch Creek and the Little Lost River valleys remain distinct for tens of kilometers into the regional aquifer. It is likely that these preferential flow zones follow high-permeability volcanic features. Flow in these zones is at a high angle to the regional hydraulic gradient. The data also delineate two regions with low isotope ratios that we interpret as relatively stagnant areas. Mixing between these stagnant areas and the regional aquifer appears to be limited. Both low-isotope-ratio zones are influenced by infiltrated water from the Big Lost River, and the western zone appears to be dominated by this water. This study highlights the potential complexity of groundwater flow patterns in a fractured-rock aquifer, illustrates the utility of isotopic data in hydrologic studies, and highlights the importance of integrating such data to maximize understanding of groundwater systems.

## APPENDIX 1. ANALYTICAL TECHNIQUES

All samples were processed under class 100 clean laboratory conditions. The water samples were weighed in the collection bottle and transferred into a weighed Teflon beaker. Sample weights were determined by difference. Overall accuracy of concentration determinations is estimated at 0.25%. The samples were spiked with $^{233}U$ tracer, which was calibrated against a gravimetrically prepared U960 standard. Spike calibrations agree to 0.05% at the two-sigma level. Samples were dried on a hot plate; the precipitated salts were dissolved in a mixture of HF, HCl, and $HClO_4$, dried, and fumed. The salts were redissolved in 7 M $HNO_3$ in preparation for anion exchange chemistry. Standard $HNO_3$ and HCL-$H_2SO_4$ anion exchange chemistry was used to purify uranium for mass spectrometric analysis.

Uranium was loaded onto outgassed Ta filaments configured in a triple filament assembly with a zone-refined Re center filament. Uranium was analyzed on a VG sector 54 mass spectrometer equipped with a WARP filter. Data acquisition was accomplished by cycling uranium masses onto the Daly knob in the mass sequence $^{233}U$, $^{234}U$, $^{235}U$, $^{235}U$, $^{234}U$, $^{233}U$. $^{238}U$ and $^{235}U$ were measured simultaneously on Faraday collectors during the cycle. Bias between the Daly and Faraday signals was determined during each cycle by comparing the $^{235}U$ signal measured in each mode. $^{233}U/^{238}U$ and $^{234}U/^{238}U$ ratios were corrected for mass fractionation by correcting the $^{235}U/^{238}U$ measured ratio to the natural value of 0.072527. Each reported isotopic measurement consisted of an average of 100 ratios, which was sufficient to achieve an internal precision of better than 0.25% for the $^{234}U/^{238}U$ ratio of most samples.

Measuring both spiked and unspiked U960 standard during the course of these measurements assessed reproducibility. $^{234}U/^{238}U$ are in excellent agreement for both spiked and unspiked standards. The mean $^{234}U/^{238}U$ is 54.86 ppm $\pm$ 0.03 ppm (two sigma). Replicates of uranium concentration and isotopic composition were also determined for sample USGS 1. The measurements are analytically indistinguishable, demonstrating excellent reproducibility of sample data.

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